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ON HIGHER CARBON AND CRUCIBLE STEELS IN SOUTHERN INDIA: FURTHER INSIGHTS FROM MEL-SIRUVALUR, KODUMANAL AND PATTINAM

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This paper touches on some further insights into the questions of the use, modes of production or antiquity of higher carbon steel or higher carbon iron alloys from studies related to some finds from surface, megalithic and early historic finds in southern India. In particular the sites discussed include the surface finds of crucible steel production identified by the author at Mel-siruvalur, finds from the megalithic burial-cum-habitation site of Kodumanal, and the early historic site of Pattinam in Kerala.

Keywords: Crucible, High carbon, Southern India, Steel.

INTRODUCTION AND HISTORY OF SOUTH INDIAN STEEL

High-carbon iron alloys are known to have been produced in Asia before they were widely used in Europe such as the traditional Indian crucible steel known as 'wootz' and Chinese cast iron. Classical Mediterranean accounts have made several references to the repute of Indian iron and steel. From the 17th century onwards European travellers have described the production of steel ingots by carburising iron, or melting constituents in crucibles in parts of South India such as Golconda/Nizamabad, Mysore and Salem in the present day states of Andhra Pradesh, Karnataka and Tamil Nadu respectively[1]. Coomaraswamy[2] (1956) has also described crucible steel processes in the Kandyan region of Sri Lanka. 'Wootz' is thought to be a European corruption of the South Indian Telegu/Kannada word for steel, *ukku*. *Ukku* may derive from the related Tamil word *'uruku'* (Tamil Lexicon)

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meaning to make molten. It could also be related to *'ekku'* the old Tamil word for sharpness/sharp spear (Tamil Lexicon; John Marr pers com.)

According to Smith[3] Indian wootz ingots, were used to forge Oriental Damascus sword found to be of a high carbon content of 1.5-2%, with a typical 'watered silk' pattern. Damascus blades, made in Khorasan and Isfahan in Persia from South Indian wootz from Golconda were thought to have been the finest weapons then made in Eurasia, reputed to cut even gauze kerchiefs[4]. The properties of South Indian steel, which became synonymous with Damascus steel, were investigated and debated during the 19th-20th centuries by several European scientists including Michael Faraday with the aim of industrial production. A typical wootz ingot analysed in 1804 by Mushet[5] had 1.3 % carbon and a dendritic structure. Studies were also prompted because the superior properties of these blades were not easily replicated with the process of forging being obscure as Indian steel rarely came unwrought to England. However investigators such as cutler Stodart were successful in replicating and forging wootz which he found to have a superior cutting edge to any other while Zschokke in 1924 found that with heat treatment this steel had special properties such as higher hardness, strength and ductility. European interest in wootz steel and Damascus blades, contributed greatly to the development of metallography in nineteenth century Europe.

Recent investigations of crucibles from surface collection from the known sites of Konasamudram, Nizamabad district, near former Golconda by Lowe[6] and Gatihosahalli in Karnataka[7] indicate specialised semi-industrial wootz processes many of which though not precisely dated may range from metallurgical activity at least over the latter medieval period i.e.16th-20th centuries. While these investigations have thrown some light on the ceramic of the crucible, studies remain to be done on the nature of the final product and metallurgical process. During the course of field investigations of copper mining and smelting in South India the author of this paper came across a previously unrecorded archaeo-metallurgical site in Mel-siruvalur, South Arcot district, Tamil Nadu, which investigations confirm to have been a production site for wootz crucible steel, by what appears to a process of carburisation of low carbon wrought iron. While some studies from this site were published in earlier papers by the author[8] a more detailed account is published here.

The find of this production centre supports the idea that wootz steel production was relatively widespread in South India, and extends the known horizons of this technology further. In addition, this paper touches upon preliminary studies on a crucible fragment excavated from the megalithic burial-cum-habitation site of Kodumanal (*c.* 3rd century BC), and reviews some reported finds from megalithic contexts and also touches briefly upon ongoing research and exciting finds of steel from the early historic site of Pattinam excavated by the author on behalf of Kerala Council for Historic Research and the megalithic site of Kadebakele (c. 700 BC) excavated by University of Michigan, University of Chicago and Karnataka Department of Archaeology.

History of South Indian steel

As for probable finds of high-carbon steel Marshall[9] reported three sword blades with 1.2-1.7% C from the Bhir mound of Taxila, from the north-western part of the Indian subcontinent (*c* 5th c. BC-1st c. AD). Greek physician Ctesia of the late 5th century BC is said to have mentioned the wonderful swords of Indian steel presented to the King of Persia. Craddock[10] points out that Greek alchemist Zosimos in the 3rd c, AD mentioned that the Indians made steel by melting soft iron in crucibles for making high quality swords.

Pliny's *Natural History* refers to the import of iron from the Seres, which could refer to the ancient Southern Indian kingdom of the Cheras, while the *Periplus of the Erythraean sea* unequivocally mentions the import of iron and steel from India[11]. Certainly trade between the ancient Cheras and the Roman world is supported by finds of late Roman coins (mid 4th c. AD) along with Chera coins from the river bed in Karur, the ancient capital of the Sangam era, while trade with *Yavanas*, i.e. Ionians or Mediterranean people, is mentioned in Sangam literature[12]. Investigations on the iron-rich megalithic sites of Tamilnadu and the Malabar (mid 1st millenium BC to early centuries AD) could be revealing. These fall within the domain and period of the Sangam Chera dynasty which might tie in with the Roman accounts of iron from the Seres, which may refer to the Chera kingdom. The moving bardic poems of the remarkable Tamil poetess Auvaiyar of the Sangam era (3rd *c.* BC-3rd *c.* AD) are particularly evocative of the skirmishes with spears between rival chieftains of that era. As such excavations at an iron age megalithic site at Kodumanal, TamilNadu (3rd c. BC), close to Karur, the capital of the Chera kingdom of the Sangam era (3rd c. BC-3rd c. AD) has revealed furnaces stacked with vitrified crucibles which were found separated from abundant iron slag[13].

Pre-Islamic Arabic literature of the 6th-7th century AD, such as Hamasa's collection of poems refers to swords of Al-Hind or *Hinduwānī* from India; while Islamic writers such as Jābīr Ibn Hayyān of the 8th century and Al-Bīrūnī of the 11th century AD make it clear that South Asian steel from India and Sri Lanka was used in many places for sword making as indicated by Bronson. The Arab Edrisi (cited by Schoff) commented that it was impossible to find anything to surpass the edge got from Indian steel. The first explicit documented evidence of the export of wootz steel from South India to make Persian Damascus blades comes from Tavernier (cited by Bronson) who in 1679 mentioned the trade in steel from former Golconda near Hyderabad, Andhra Pradesh which was the only sort which could be damascened by Persian artists, by etching with vitriol.

Past observers of the manufacture of wootz steel in India have commented on the process of carburisation of iron to steel in crucibles where a batch of closed crucibles with the low carbon iron charge were stacked in a large furnace and fired in a long 14-24 hour cycle at high temperatures of not less than 1200^{0}C in a strongly reducing atmosphere[14]. Three different types of crucible processes have been described by nineteenth century travellers varying from region to region ie. the Deccani or Hyderabad process, the Mysore process and the Tamil Nadu processes. In the Tamil Nadu process and the Mysore process, the charge consisted of wrought iron or bloomery iron (i.e. low carbon iron with no more 0.1% C) produced separately which was then stacked in closed crucibles and carburised in a large furnace[15]. But while the Mysore process charged the wrought iron with carbonaceous matter, Wood's observations on crucible processes in Salem and Arcot districts in Tamil Nadu suggest that only iron was charged and the crucible containing the ingot was not fast cooled in water as in the Mysore process. The Deccani process from the Hyderabad/Golconda region was renowned for the best quality wootz from where the best known site is Konasamudram. One of the processes followed here was not of carburisation of a wrought iron bloom but of fusion of two separate pieces of cast iron (i.e. high-carbon iron of about 4% C) and a low carbon iron bloom so producing an alloy of intermediate composition[16].

The known sites of crucible steel production in South India i.e at Konasamudram and Gatihosahalli date from at least the late medieval period, i.e. 16th century. But, although these may be earlier, systematic excavations have not yet been carried out on sites of crucible steel production to determine their antiquity. The existing research on wootz steel at these sites has been more concerned with metallurgical re-construction of the wootz process based on surface finds. The investigation presented here is also from undated surface finds at a mound in Mel-siruvalur village, South Arcot district in Tamil Nadu.

Mel-siruvalur: Archaeometallurgical Evidence for High-carbon Crucible Steel Processing

In November 1991 Srinivasan made field investigations (with the help of the State Department of Mines and Geology, Madras, and Geological Survey of India-Madras Circle) of old copper workings in Mamandur, South Arcot district, about 40 km from the nearest town of Tiruvannamalai. While searching for evidence of copper smelting activity the author came across metallurgical debris near the village of Mel-siruvalur about 5 km from Mamandur, which is shown further to be related to crucible steel production. Evidence of metallurgical activity came from a mound just behind the village of about 25 m x 8-9 m wide and upto 5 m high and from some trenches near the houses. The villagers had no memories related to the metallurgical activity. Occupation of the area in antiquity is indicated by pottery sherds collected adjacent to an old canal, about 1/2 a km away from the mound. Slag debris and crucible fragments were also found all around the canal site. Among the sherds were many large rim fragments, about 3cm thick, belonging to huge storage jars about 60 cm in diameter. These had no slip, and were found to be tempered with rice hulls. Late C.S. Patil (pers. comm.) of the Mysore Archaeological Survey has pointed out their resemblance to megalithic storage jars of red ware without slip. The megalithic occupation in Tamil Nadu, which begins in the 1st millennium BC, continues to as late as the 5th century AD. Megalithic dolmens have been found in Thiruvannamalai and Tirukoilur taluks in South Arcot district. Sasisekaran also observed signs of megalithic occupation at the site. Also found were pottery sherds of painted ware with a red slip, decorated with a chain or hatched design, which were identified by late C.S. Patil as being of the late medieval period (*c*. 16th century AD). Without more detailed survey and investigations, the relation of these pottery assemblages to the metallurgical activity cannot be ruled out. In cross-section it could be seen that the pottery was re-inforced with rice hulls. Interestingly, several hollow tapering terracotta jars about 70 cm long were also found stacked along the walls of the canal although their function could not be determined (Fig. 1). They did, however, resemble the legs of megalithic sarcophagi, while large jar fragments are also known from Indo-Roman contexts.

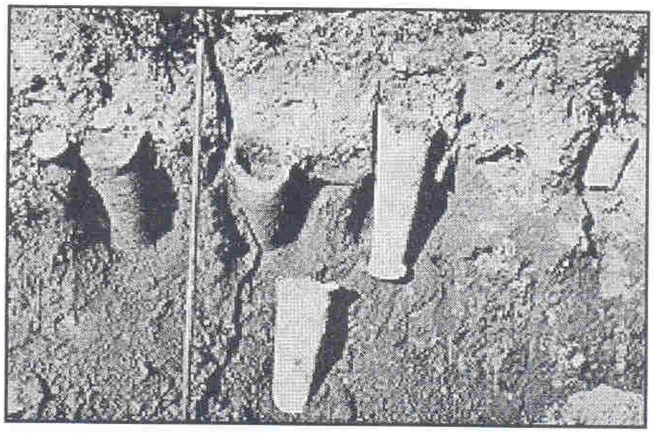

Fig. 1. Large tapering jar-type fragments at Mel-siruvalur.

Description of Archaeo-metallurgical Debris at Mel-siruvalur

Numerous crucible fragments were found at the Mel-siruvalur mound together with fragments of glassy slag, charge and debris (Fig. 2). When re-constructed the fragments of crucibles resembled aubergine-shaped closed crucibles used for wootz steel production known from other sites in South India such as Gatihosahalli and Konasamudram, indicating that the crucibles had been broken to retrieve the ingots. Thick covering lids of a diameter of about 7 cm, which would have sealed the refractory vessel during firing with the iron charge, were found. The exterior surface of the crucibles was covered with thick black ash glaze indicating that the crucibles were fired under highly reducing conditions. That the crucibles had been stacked together could be discerned from the flat vertical ridges seen along some crucibles where they were pressed against other crucibles. The base of the lid was uneven and bore imprints of a fibrous pattern suggesting that the crucible could have been stuffed with leaves and grass as carbonaceous material for carburising the charge.

Pieces with interior glazed surfaces and distinctive concave 'fins' of glassy slag which formed a ring around the middle of the crucible were found (Fig. 2). The presence of fins of glassy slag indicate that the charge was in a molten state and that some slag molten separated and floated on top of the molten metal. The curvature of the slag fins suggests that the liquid slag filled the space between the convex meniscus of the molten charge. Several curved bases of the crucibles, about 0.8-1.5 cm thick were among the finds. A lining of glassy slag with a honeycomb pattern covered the area below the fin to the crucible base which would have been occupied by molten charge and solid ingot. Some of the slag linings on the crucible bases had remnants of the rusty charge attached to them. The dimensions of the various fragments indicated that the ingots were of a diameter of *c.* 2.5 cm. Rusty patches and rusty globules could be seen scattered on the ring of slag fin, above this fin, along the crucible walls and even below the lid. This must have been caused by splashing of molten charge which could happen due to a carbon boil reaction. All of the above suggests that the ingot solidified from a molten charge.

About 70 m away from the mound were a set of two trenches inter-connected in a pinch and swell shape of about 10 m long. One of these was clearly the furnace area, as it contained several tapering tuyeres fragments with an inner diameter *c.* 1.5 cm, and varying from 0.8-2 cm thick, along with furnace remnants, consolidated mud and slag. The other trench contained

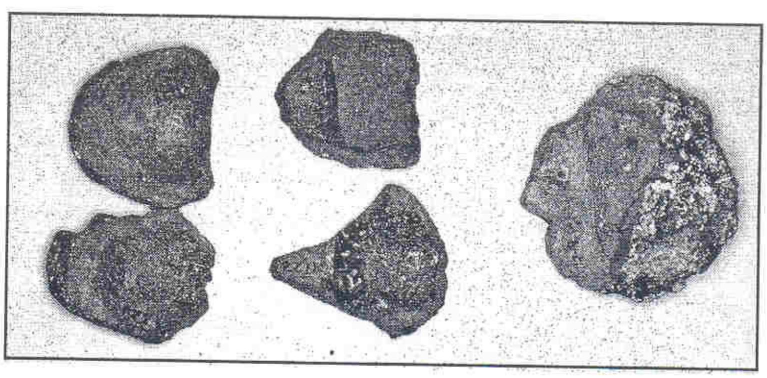

Fig. 2. Assemblage of steel-making crucible fragments from Mel-siruvalur.

only blocks of slag 20 cm high and 20 cm in diameter with a flow texture; indicating that it had been used to tap out slag from the main furnace. Slag specimens from these areas were also collected.

Analytical Results and Discussion

The blackish glaze on the exterior suggested that the crucibles were fired under highly reducing conditions at high temperatures with heavy attack by fuel ash. This glazed exterior was seen to be impregnated with several large and small white inclusions which were not found in the interior cross-section. SEM/EDS analysis of a fragment of white inclusion showed it to be quartz (Si ≈ 47%, 0 ≈ 53% normalised) (Table 1). Thus the crucible exterior seems to have been rolled in crushed quartz (SiO_2) to protect the exterior of the crucible from melting down from attack by fuel ash and associated alkaline fluxes since the silica would make it more refractory and raise melting point.

Table 1. Some SEM/EDS* analyses of constituents in sections of fragments of crucibles from Mel-siruvalur: constituents examined in section of glassy slag lining on interior of crucible.

Constituent	Fe	Al	Si	Ca	K	P	S	Ti	Cu	O	Total
Prill	99.94	0.03	1.82	0.08	0.0	0.14	0.02	0.0	0.0	-	102.03
Crucible cross-section of 86 mm	4.95	11.89	34.96	28.06	2.92	1.37	8.36	7.31	0.17	-	99.99
Area within rice hull (spot)	2.94	3.73	10.05	0.46	0.48	0.0	0.01	0.25	0.06	-	17.98
Area around rice hull 1 (spot)	0.09	0.0	38.90	0.0	0.0	0.01	0.0	0.0	0.0	-	39.00
Area around rice hull 2 (spot)	0.13	0.0	38.74	0.0	0.0	0.10	0.0	0.0	0.0	-	38.97
Glassy matrix within crucible	5.80	7.40	22.99	11.23	3.36	0.27	0.02	0.77	0.03	-	51.87
Slag fin	8.47	6.53	14.47	5.64	1.60	0.41	2.52	13.0	0	-	52.64
Glazed exterior	2.78	5.90	15.16	3.82	2.35	0.06	0.02	0.59	0.0		30.68
White inclusions in glazed exterior	0.17	0.0	46.60	0.0	0.03	-	0.01	7.00	-	-	53.81

*SEM/EDS analysis was done using HITACHI S-570 with link AN-1000 with ZAF correction with instrumental accuracy of around 8% at 25%. Lighter elements such as O, C, N etc. not analysed for.

The fabric of the crucible was also black, porous, very carbonaceous with tiny pieces of charcoal intact and also consisted of a glassy network with distinctive coked rice hull relics dispersed in the matrix, the interiors of which were either voids, had charred carbonaceous remains or fused glassy remains. The carbon-rich crucible strongly suggests that the process followed was of carburisation of wrought iron charge. The inclusion of rice hulls in the refractory material has been noted in the manufacture of Deccani wootz crucibles by Lowe[18]. Lowe (ibid.) postulates that these were added for their high silica and carbon content, making the crucible a particularly effective re-inforced composite refractory material; both to withstand very high temparatures over a very long firing cycle and to maintain a highly reducing environment to enable carburisation of the iron charge. It may be significant that SEM/EDS analysis of the fused networks just around, or circumscribed, by the rice hull relics showed them to have a fixed composition of 38-39% Si (Table 1). Since it is not possible to analyse for carbon by SEM, the probability remains that the rest is carbon, which may suggest that the compound is silicon carbide (SiC_4) which would have about 37 wt. % Si, while silica (SiO_2) would have closer to 47%. In glassy matrix at large the presence of both silicon of up to 23% and aluminium of 6-7% is seen (Table 1). Lowe, Merk and Thomas[19] have shown that wootz refractories from Konasamudram were reinforced by the formation of fibres of the alumino-silicate mullite which is a strengthening material in the field of high-performance ceramics. The practice of inserting rice hulls to re-inforce the fabric of pottery is also seen at this very site in the pottery mentioned earlier.

Sections for microscopy were taken from different fragments of crucibles representing the lid, the glassy fin and the crucible base. The crucible base had a thin layer of glassy slag with the honeycomb pattern with some remnant rusty charge. The cross section indicated that the rusty layer formed due to the molten charge was extremely thin, no more than 10 μm thick. Although tiny uncorroded prills were detected under optical microscopy in this rusted layer following sectioning, by the time fine polishing was done they had fallen out of the loose rust matrix. Uncorroded metallic prills were best detected trapped and preserved in the glassy slag fins and slag coating on the interior of the crucible, and the exterior black glaze in the lid section. While the prills in the exterior glaze may be attributed to factors such as the contamination of particles of charge or iron filings which would have also been carburised or the splashing of molten metal, the prills that were found trapped within the glassy slag fin and lining of the interior of the crucible can be convincingly linked to the final composition of the molten charge. The prills were tiny of a diameter of less than 100μ, with most prills in the glassy slag on the interior of the crucible being no more than 10-20μ. Analyses of the prills using Electron Probe Microanalysis with wavelength dispersive X-ray spectrometry (EPMA-WDS) confirmed that they are steel prills (Table 2, 3).

Table 2. Mel-siruvalur crucible: EPMA (wt%) analysis of prills in a section of a lid fragment*

Constituent	Fe (wt %)	Cu(wt %)	As(wt %)	S(wt %)	Total(wt %)
Prill 1	97.615	0.038	0.023	0.565	98.241
Prill 2	102.385	0.075	-	0.251	102.711

*Analysed by EPMA on a polished and carbon-coated cross-section using JOEL Superprobe JXA-8600 at 20 KV with ZAF correction, within instrumental accuracy of 1% over 100% using probe diameter of 50μ. Trace Si and P were noted but were not analysed by EPMA; however these elements were less than or around 1% when seperately analysed by using SEM with EDAX analysis (HITACHI S-570 with link AN-1000).

Table 3. EPMA-WDS (wt%) analysis of constituents in glassy slag fin on interior of crucible#

Constituent	Fe	Al	Si	Ni	Cu	Ti	P	S	Ca	Total
Prill	82.28	0.18	3.25	1.74	0.27	0.09	9.95	0.12	1.35	99.23
Glassy slag	0.32	8.62	24.46	0.0	0.0	0.89	0.02	0.0	7.13	41.44

#Analysed by EPMA on polished, carbon-coated cross-section using JOEL Superprobe JXA-8600 at 20 KV with ZAF correction, within instrumental accuracy of 1% over 100% with probe diameter of 5μ. Lighter elements such as O, C, N etc. not analysed for.

The etched microstructure of the largest detectable prill of all (Prill 1) is shown which was found in the glazed exterior of the section of the lid of a crucible. It has a lamellar structure of fine pearlite eutectoid (of equilibrium composition of 0.8% C) inside original hexagonal grains of austenite. This suggests that it derives from a very good quality hypereutectoid high-carbon (>0.8%) steel. The prill had a hardness of around 400 VPN which is within the range for normalised steel of *c.* 1% carbon, suggesting that the crucible had not been quenched. The presence of much smaller amounts of a light unetched network between grains near the boundaries and as occassional needles in the pearlite was also noted; SEM-EDAX analysis suggested that this was mainly cementite (iron carbide) with some phosphorus impurities; ie. cementite-phosphide. Iron phosphide tends to form a ternary eutectic of steadite along with pearlite and its presence could indicate a slightly higher carbon content of about 1%-1.2%. Smallers prills in the lid also had a similar micro-structure.

It is the prills trapped in the glassy slag fin of 3-4 mm thick (Fig 3, 4) and slag lining of 1 mm thick of the interior of the crucible that best reflect the composition of the molten liquid from which the ingot solidified. These also have the same structure as described above of lamellar pearlite eutectoid inside hexagonal grains surrounded by a intergranular network of cementite with some cementite needles. Table 1 also indicated the presence of phosphorus in the cementite network. Hence these strongly suggests that the final product or solidified ingot was a high-carbon steel of around or over 1% C. The micro-structure of the prills compares well with that of a 1.2% C steel illustrated in Avner[20]. The impression of the honey-comb pattern in the slag lining and fin may also be related to the two-phase micro-structure of the molten metal with the carbide network surrounding the polygonal grains of high temperature austenite which forms pearlite at lower temperatures.

Of the prills, one somewhat larger prill in the exterior glaze of the crucible, has a structure which slightly differs from the rest and shows an excess of cementite and steadite compared to the other prills with the pearlite colonies getting more spheroidised, indicating a slightly higher carbon content than the others of up to 2% C. This may be due to fact that the exterior of the crucible was in contact with excess carbon from the fuel, perhaps getting over-carburised, while another possibility is that the exterior of the crucible was packed with crushed crucible fragments as referactory from a previous process. While it would be expected that by the end of the process, any iron existing in-situ in the crucible prior to the process would get carburised to the same equilibrium conditions, there may small variations in the reducing/ carburising conditions in different parts of the crucible.

Qualitative analysis of a few samples of the slag collected from the second trench by SEM/EDS showed that the major constituents were iron and silicon suggesting that these may be fayalite (iron silicate) type iron

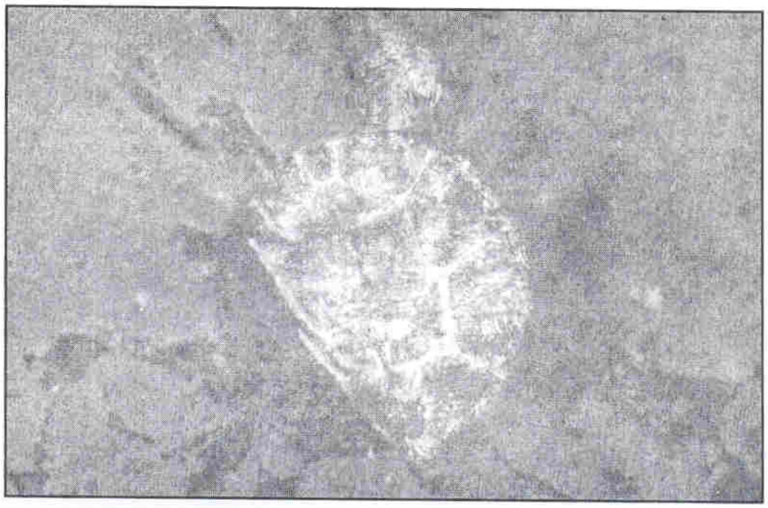

Fig. 3. Mel-siruvalur crucible: High-carbon steel prill (1.2% C)
(Of dimension 50 microns at 400X).

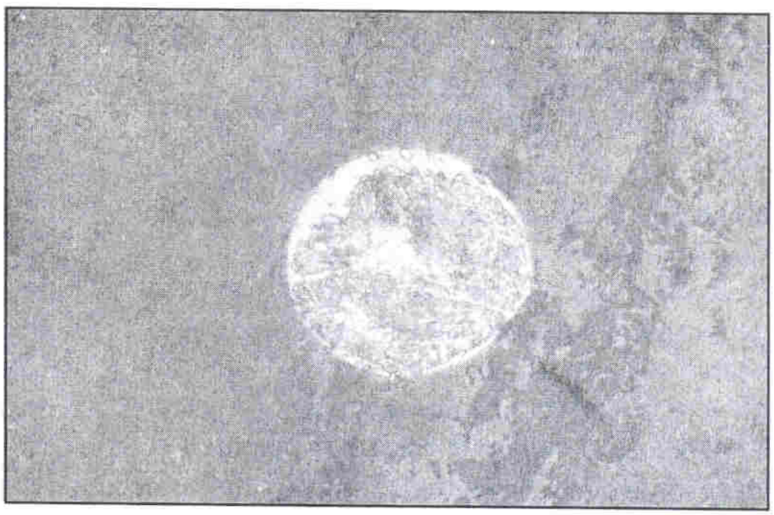

Fig. 4. High-carbon steel prill in Mel-siruvalur crucible
(of diameter 10 microns) (1000x).

slags. Hence it appears that the iron charge was being smelted by the bloomery process in the trenches. The low carbon iron bloom produced here may have formed part of the charge to produce high carbon iron by the wootz crucible process in the area where the mound with the crucibles was found.

Megalithic Burial-cum-habitation Site of Kodumanal Dating Back to 300 BC

Kodumanal (11^0 6′ 42″ N 77^0 30′ 51″ E) in Tamil Nadu is perhaps one of the most important sites in terms of providing what may be the earliest known evidence for the use of crucible methods in ferrous processing. Here an impressive megalithic burial cum habitation site dating back to 300 BC was uncovered which showed evidence of considerable semi-industrial activity with remnants ranging from iron, steel, copper artifacts, gem stone cutting, polishing and bead making and semi precious stone working including quartz, beryl etc. The site Kodumanan finds mention going back to classical Tamil literature of the Sangam era. The Tamil Sangam poems of Patirrupattu refers to the export of goods from Kodumanal to the Roman world.

Iron artifacts found at Kodumanal included tanged and barbed arrowhead, fragments of rings, chisels, battle-axe, swords, daggers, horse stirrup, leaf-shaped arrowheads, iron bead, an iron bell, sickles, hooks etc[21]. The length of swords in the burial complex varied from 120-150 cm. One of the finds included a chisel with an edge of higher carbon steel (about 1% carbon)[22].

At Kodumanal fifteen trenches were laid 300 m north of the iron furnace area and this yielded two crucible furnaces of which one was found in an unused condition. These furnaces were at a depth of 125 cm below the ground. At least 12 small furnaces surrounded the main crucible furnace. The main furnace was oval measuring 112 cm in the north south direction and 100 cm east west. The depth of the furnace was 40 cm. The furnace wall was made of burnt clay with a thickness of 20 cm. The burnt clay had a rectangular hole made in an acute angle. The small circular furnaces surrounded the main furnace were 30 cm in diametre at the mouth with a small hole of depression in the center and were connected to the main furnace through burnt clay pipes. A broken vitrified crucible fragment found *in situ* in the small furnace was technically examined by this author as reported further (Fig. 5).

This broken fragment from Kodumanal was distinctive from two small bowl-like open crucibles which were also found at the site which may have been used to cast precious metal. This broken piece was blackened and vitrified, rather more like the crucibles related to steel making from Mel-

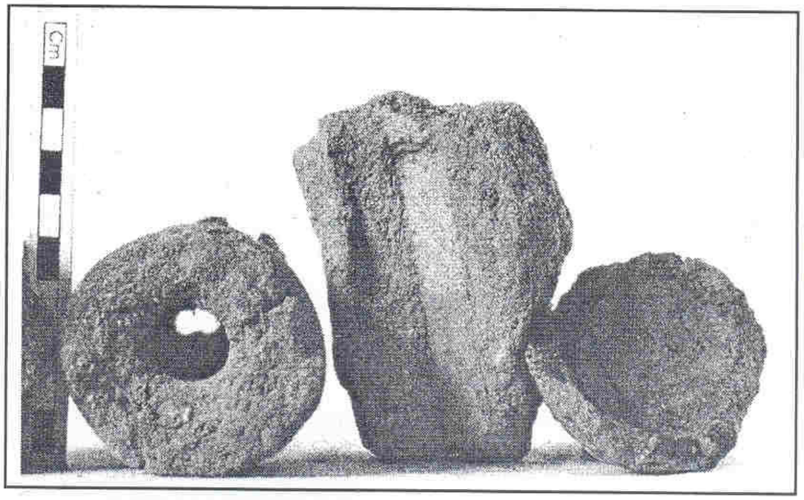

Fig. 5. Kodumanal (c. 3rd century), crucible, tuyere and other debris from ferrous processing.

siruvalur, although very much more friable suggesting that it was not as highly refractory. Significant amounts of iron and silicon were detected in the overall crucible fabric. Although prills have yet to be identified, no significant amounts of other metals such as copper, gold, silver, lead etc. were found which may have been expected if they were related to a non-ferrous melting process. The fact of finding certain small regions which seem to be especially iron-rich might be significant in terms of evidence suggesting that it was used for ferrous metal processing. Like the Mel-siruvalur fabric there were inclusions of pure silicon, i.e. quartz fragments. Trace elements of titanium are found in the iron-rich regions of both crucibles. Although titanium is a common iron impurity, nevertheless this may not be without significance, since titanium is a grain refiner which reduces grain size and improves toughness of microalloyed steels. The role of micro-alloying constituents of vanadium in promoting the formation of carbide-banding responsible for the pattern in the forged wootz ingots has been argued by Verhoeven[23]. Fig. 6 is an elemental distribution map for iron in a polished and carbon-coated specimen of the crucible fragment from Kodumanal, obtained from an x-ray area scan prepared with the characteristic x-ray signals of iron derived from a wavelength dispersive x-ray spectrometer (EPMA-WDS, using JOEL Superprobe JXA 8600 at 20 kv), indicating iron-rich areas.

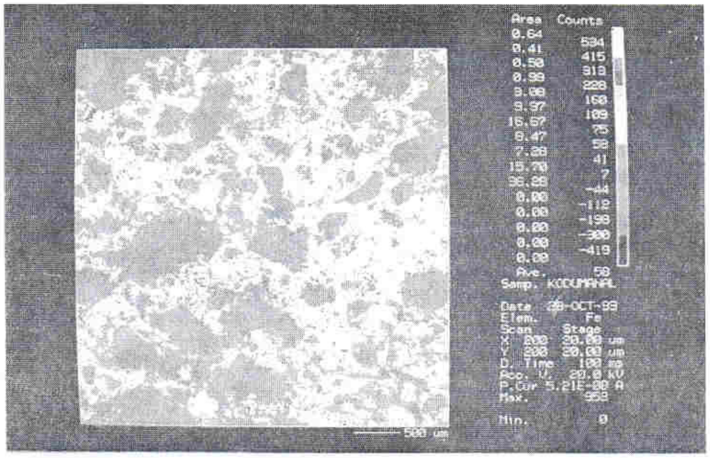

Fig. 6. Kodumanal crucible fragment: Elemental distribution map obtained from an x-ray area scan using EPMA-WDS.

Table 4. SEM/EDS semi-quantitative investigations on a crucible fragment from megalithic excavation of Kodumanal (c. 3rd century BC) given to Srinivasan by Rajan of Tamil University, Tanjavur

Sr No.	Fe (wt %)	Si (wt %)	Al (wt %)	Na (wt %)	Ca (wt %)	Ti (wt %)	Cu (wt %)
Whitish patch in crucible	50.07%	30.78%	14.5%	1.4%	1.43%	0.66%	0.4%
Matrix of crucible	14.28%	28.20%	16.64%	1.21%	3.36%,	0.46%	0.09%
Small whitish patch	80.25	41.96	30.87	2.61	1.51	0.7	-
Bright spot (quartz fragment)	0.37	80.28	-	0.14	-	0.05	-

Note: Given that the crucible was quite friable with porosities and the whitish patches very small it was not easy to get a very accurate analysis and hence results shown are not normalised to 100%. Analysis was done using HITACHI S-570.

Review of Some Megalithic Sites where Higher Carbon Steel Finds are Reported

Intriguing investigations were reported by Mudhol on specimens of a set of two javelins and one dagger from megalithic burials excavated from Andhra Pradesh from the sites of Uppalapadu and Chinnamarur, Mahaboobnagar district and Kadambapur, Karimnagar district of Andhra Pradesh. Although completely corroded these were probably of high-carbon steel from the reliced pseudomorphic structures and carbon content of between

1.6-1.8% carbon. As for the dates of the sites, few carbon dates are available from Andhra Pradesh. Charcoal from Dharnikota near Amaravati, on the bank of the Krishna gave dates of 475±100 BC and 390±100 BC for the early historic phase that succeeded the megalithic culture. A time bracket of 6th century BC to 2nd century BC was broadly attributed to the active megalithic period in Andhra Pradesh by Rao[25].

Table 5 Some megalithic steel specimens reported in Mudhol[24]

Source	Mudhol 1997: 62	Mudhol 1997: 64	Mudhol 1997: 63
Object	Javelin, Uppalapadu, AP	Javelin, Kadambapur, AP	Dagger piece, Chinnamarur, AP
Condition of the object	Highly corroded and fragile	Highly corroded and mineralized	Completely mineralized
Method of analysis	-Carbon measured in a eudiometer by direct combustion -Details of other analyses not available	-Carbon measured in a eudiometer by direct combustion -Details of other analyses not available	-Carbon measured in a eudiometer by direct combustion -Details of other analyses not available
Iron	Not measured	Not measured	Not measured
Carbon	1.64%	1.76%	1.8%
Phosphorus	0.289%	0.296%	0.214%
Sulphur	0.062%	0.05%	0.045%
SiO_2	5.02%	11.3%	16.04%
Al_2O_3	0.64%	0.92%	0.48%
MgO	0.08%	1.89%	0.64%
CaO	0.17%	3.96%	0.69%
Fe_2O_3	73.7%	61.83%	64.63%
FeO	6.06%	6.52%	4.86%
LOI	10.4%	8.26%	9.38%
Micro-structural features	Network of psuedomorphic structures of relic carbide particles and spiny pseudomorphs of cementite reported.	Pseudomorphic pearlite structure and few spheroidized relic carbide particles, with deep furrows	No metallic structure but white metallic looking cementite particles detected
Conclusions	Structure is thought to have been related to a rapidly cooled high-carbon steel	Object is thought to have been made of welded highly carburised wrought iron strips and fabricated at over 800^0 C	Object is thought to have been high-carbon steel, with presence of cementite prior to forging

ONGOING RESEARCH IN PATTINAM BY AUTHOR OF EARLY HISTORIC IRON AND MEGALITHIC FINDS OF STEEL

The author in collaboration of Kerala Council for Historic Research is hoping to embark on a project concerning metallurgy of ferrous metal artefacts excavated from the early historic Indo-Roman settlement of Pattinam, in Kerala. This site is being excavated by Kerala Council of Historic Research under Cherian, Selvakumar and Shajun. It is thought that this site may coincide with the Muziris of Roman accounts as an important trading emporium on the coast of Kerala. As mentioned before, one theory has been that the export of iron from the Seres in Pliny's *Natural History* may refer to the Chera region covering parts of Tamil Nadu and Kerala. In that connection it may be significant to mention that the micro-structure of an iron nail fragment examined by the author showed a clear structure of a higher carbon steel, from the micro-structure apparently approaching the pearlitic eutectic composition of 0.8% (Fig. 7). On the other hand, the micro-structure of a Roman nail, from a cache of Roman nails in Perthshire published in Scott[26], showed it to be of pure alpha phase wrought iron with slag stringers and no traces of carbon. This and the literary accounts mentioned before suggests that the higher quality higher-carbon steel was of local south Indian origin.

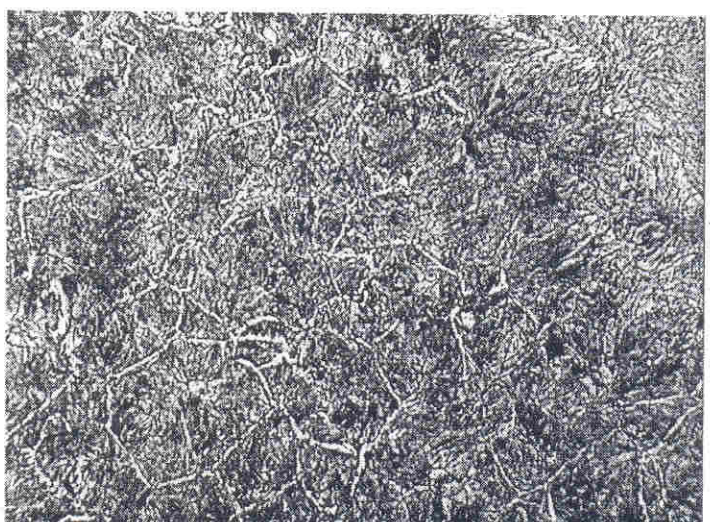

Fig. 7. Pattinam: nail showing almost purely pearlitic structure (0.8% steel).

The author has also visited and been studying material from the iron age site of Kadebakele, which is on the outskirts of the 14th-16th century world heritage site of Vijayanagara in Karnataka close to the village of Anegondi overlooking the Tungabhadra river. This site has been excavated by a collaborative team from the University of Michigan (headed by Prof. Carla Sinopoli), and the University of Chicago and the Karnataka Department of Archaeology,. The site was occupied from at least the early first millennium BC into the early centuries AD. Excavations revealed numerous iron artefacts. Carbon dates from the Iron Age or 'megalithic' period span from c. 800-400 BC. Some specimens examined had structures of higher carbon pearlitic steel [presented in paper read at 6th International Conference of Beginning of the Use of Metals and Alloys (BUMA-6) by S.Srinivasan, C. Sinopoli, K. Morrison, K. Gopal and S. Ranganathan].

General Discussion

The preliminary investigations reported here indicate that crucible steel production was carried out, in the pre-industrial era, at a hitherto unreported site at Mel-siruvalur, South Arcot district, Tamil Nadu. Analytical investigations indicate that the process can be re-constructed as the carburisation of a low carbon iron blooms in crucibles by firing them under carbonaceous and highly reducing conditions. There is evidence for the manufacture of iron blooms in the vicinity which may have provided the charge along with carbonaceous organic matter. Microstructural, micro-hardness and EPMA & SEM examination of etched sections of a lid fragment, glassy slag fins and slag lining (adjacent to the molten ingot from two crucible fragments) showed a number of prills with the majority having a structure akin to a hypoeutectoid carbon steel with 1-1.2% C. The general uniformity of the micro-structure of prills indicates the final product had a uniform composition and that the wrought iron charge was heated to the molten state before being carburised. Indeed Rostoker and Bronson[27] have pointed out that it is the melting of wrought iron in the presence of solid carbon which would result in a homogenous steel, rather than the processes of cementation of solid wrought iron with gaseous carbon, or the process of allowing the solid metal to come in contact with liquid cast iron. In comparison the process of crucible steel manufacture from the Islamic site of Merv, Turkamenistan is thought to have been of co-fusion of cast iron and wrought iron, and the prills were noted to have a range of compositions from cast iron to steels of 0.1-0.8% C[28].

The micro-structure of the prills etched in nital showed a structure that can be attributed to a hypoeutecoid steel with 1-1.2% C with dark lamellar pearlite eutectoid inside original hexagonal grains of austenite, surrounded by a network of unetched light cementite/cementite-phosphide. The watered silk pattern on Damascus swords basically consists of lamellar darkly-etched high carbon pearlitic steel interwoven with a network of lightly-etched iron carbide or cementite/cementite-phosphide: formed by the forging of a high carbon iron ingot followed by etching. Studies by Wadsworth and Sherby[29] on simulation experiments for making steel for 'Damascus' blades used steel with 1.7% C which had a similar micro-structure of pro-eutectoid cementite surrounding prior austenite grain size surrounding pearlite colonies to reproduce the watered silk pattern. According to their findings steels of such compositions were confirmed to be readily formed at around 800-900^0 C, with the formation of globular dispersions of the carbides. The structure of a modern razor blade with 1% C illustrated in Rostoker and Bronson[30] is also somewhat similar, indicating that such a composition of steel can give a sharp cutting edge. Gilmour[31] has pointed out that it is the segregation of phosphorus in the lightly etched pattern, i.e. the carbide network., that helps to retain the watered silk pattern since it prevents migration of the carbon. Indeed the presence of phosphorus has been detected in the cementite network in the Mel-siruvalur prills. Thus Mel-siruvalur seems to have been a site for manufacturing 'wootz' ingots with around 1-1.2% C of the type that could have been used for making 'Damascus' steel or watered steel. In a small shop in Ootacumund, in Tamil Nadu state of India, Srinivasan came across a watered steel sword with a hilt made of 'bidri' i.e. the late medieval silver damascened zinc alloy typical of the old Bidar district very near Hyderabad (former Golconda) in South India, which suggested that such watered steel blades could also have been made near the production sites of wootz steel.

To make the wrought iron molten, the charge would have had to have been heated to about 1400^0 C while the highly reducing conditions may have helped to reduce melting point a bit. The crucibles were designed to withstand such temperatures with the exterior surface being packed with quartz to

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